

## Toward accurate CO<sub>2</sub> and CH<sub>4</sub> observations from GOSAT

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Received 21 April 2011; revised 14 June 2011; accepted 20 June 2011; published 30 July 2011.

[1] The column-average dry air mole fractions of atmospheric carbon dioxide and methane ( $X_{\text{CO}_2}$  and  $X_{\text{CH}_4}$ ) are inferred from observations of backscattered sunlight conducted by the Greenhouse gases Observing SATellite (GOSAT). Comparing the first year of GOSAT retrievals over land with colocated ground-based observations of the Total Carbon Column Observing Network (TCCON), we find an average difference (bias) of  $-0.05\%$  and  $-0.30\%$  for  $X_{\text{CO}_2}$  and  $X_{\text{CH}_4}$  with a station-to-station variability (standard deviation of the bias) of  $0.37\%$  and  $0.26\%$  among the 6 considered TCCON sites. The root-mean square deviation of the bias-corrected satellite retrievals from colocated TCCON observations amounts to 2.8 ppm for  $X_{\text{CO}_2}$  and 0.015 ppm for  $X_{\text{CH}_4}$ . Without any data averaging, the GOSAT records reproduce general source/sink patterns such as the seasonal cycle of  $X_{\text{CO}_2}$  suggesting the use of the satellite retrievals for constraining surface fluxes. **Citation:** Butz, A., et al. (2011), Toward accurate CO<sub>2</sub> and CH<sub>4</sub> observations from GOSAT, *Geophys. Res. Lett.*, 38, L14812, doi:10.1029/2011GL047888.

### 1. Introduction

[2] Space-based remote sensing of the CO<sub>2</sub> and CH<sub>4</sub> column-average dry air mole fractions ( $X_{\text{CO}_2}$  and  $X_{\text{CH}_4}$ ) has the potential to provide global observational constraints on CO<sub>2</sub> and CH<sub>4</sub> fluxes across the surface-atmosphere boundary and to foster insight into the related biogeochemical cycles. However, measurements of  $X_{\text{CO}_2}$  and  $X_{\text{CH}_4}$  face challenging accuracy requirements. The target accuracy for regionally and weekly averaged  $X_{\text{CO}_2}$  and  $X_{\text{CH}_4}$  is on the order of a few tenths of a percent out of background concentrations of roughly 385 ppm (parts per million) and 1.8 ppm, respectively, where

the requirement for  $X_{\text{CH}_4}$  is somewhat less stringent than for  $X_{\text{CO}_2}$  [e.g., Chevallier et al., 2007; Meirink et al., 2006].

[3] Currently, the SCanning Imaging Absorption spectroMeter for Atmospheric Cartography (SCIAMACHY), in orbit since 2002, and the Greenhouse gases Observing SATellite (GOSAT), in orbit since January 2009, aim at achieving this goal by exploiting absorption spectra of backscattered sunlight in the shortwave-infrared (SWIR) spectral range. SCIAMACHY has been shown to provide accurate  $X_{\text{CO}_2}$  [e.g., Reuter et al., 2011] and  $X_{\text{CH}_4}$  [e.g., Frankenberg et al., 2005] if lightpath modification due to scattering by atmospheric particles is taken into account by the retrieval methods. First retrievals from GOSAT have been reported by Yoshida et al. [2011] and Morino et al. [2011]. They use relatively weak CO<sub>2</sub> and CH<sub>4</sub> absorption bands ( $\sim 6100 \text{ cm}^{-1}$ ) in combination with the O<sub>2</sub>A-band ( $\sim 13,000 \text{ cm}^{-1}$ ) to retrieve  $X_{\text{CO}_2}$ ,  $X_{\text{CH}_4}$ , and simultaneously the amount of a predefined aerosol type in the boundary layer. Validation of their results by ground-based measurements of the Total Carbon Column Observing Network (TCCON) [Wunch et al., 2011] shows a standard deviation of the differences of generally more than 1%, which is too large to unambiguously identify characteristic source/sink patterns such as the seasonal cycle of  $X_{\text{CO}_2}$ . This shortcoming might be partly due to lightpath modification not only depending on the particle amount but also on particle size and height.

[4] Here we demonstrate that we can deliver retrievals of  $X_{\text{CO}_2}$  and  $X_{\text{CH}_4}$  from GOSAT with improved quality and that timeseries of individual GOSAT retrievals allow for identifying characteristic source/sink patterns. This is achieved using the O<sub>2</sub>A-band, the weak CO<sub>2</sub> and CH<sub>4</sub> absorption bands around  $6100 \text{ cm}^{-1}$ , and the strong CO<sub>2</sub> and H<sub>2</sub>O absorption bands around  $4850 \text{ cm}^{-1}$  in order to simultaneously retrieve information on the particle amount, size, and height, together with  $X_{\text{CO}_2}$  and  $X_{\text{CH}_4}$ . The quality of the retrieval results is evaluated through comparison with coinciding ground-based observations at 6 TCCON sites.

### 2. GOSAT Observations and Auxiliary Data

[5] The Thermal And Near infrared Sensor for carbon Observation (TANSO) - Fourier Transform Spectrometer (FTS) onboard GOSAT observes sunlight backscattered by the Earth's surface and atmosphere in 3 channels covering the SWIR spectral range. A further channel records thermal radiation in the longwave infrared, which is not used here. Over land, the instrument collects light about the nadir ( $\pm 35^\circ$  across track,  $\pm 20^\circ$  along track). Over the ocean, it also looks at the glint spot, the point of specular reflection at the water

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surface. The FTS has a maximum optical path difference of 2.5 cm and an instantaneous field-of-view of 15.8 mrad mapping into a spectral resolution of  $\sim 0.3 \text{ cm}^{-1}$  and a circular footprint with  $\sim 5 \text{ km}$  radius at the sub-satellite point. Backscattered sunlight is recorded in 2 orthogonal polarization directions from which we calculate the total backscattered radiance (Stokes parameter  $I$ ) as suggested by Yoshida *et al.* [2011]. The radiometric calibration of the spectra is based on the Mueller matrix calculus of Kuze *et al.* [2009] and the pre-launch measured calibration data (with corrections) available from GOSAT's instrument support. The latter also provides the tabulated instrument line shape (ILS) used by our algorithm. The TANSO-Cloud and Aerosol Imager (CAI), the other instrument onboard GOSAT, delivers cloud flags for several hundred ground pixels within a single TANSO-FTS footprint which we use for cloud screening (see auxiliary material for details).<sup>1</sup>

[6] Further auxiliary information necessary to accurately retrieve  $X_{\text{CO}_2}$  and  $X_{\text{CH}_4}$  is collected by a preprocessing step. Vertical profiles of pressure, temperature, and humidity as well as surface pressure and wind speeds are extracted from the ECMWF (European Centre for Medium-Range Weather Forecasts) ERA-Interim analysis provided 6-hourly on a  $1.5^\circ \times 1.5^\circ$  latitude  $\times$  longitude grid. ERA-Interim fields are interpolated to the time and center location of the TANSO-FTS footprint taking into account surface elevation and its variability through the GTOPO30 database. The initial guess for the CO<sub>2</sub> and CH<sub>4</sub> vertical concentration profiles is taken from CarbonTracker [Peters *et al.*, 2007] and TM4 [Meirink *et al.*, 2006] model runs for the year 2008 and 2007, respectively.

### 3. Retrieval Method

[7] The retrieval method has been described in detail by Butz *et al.* [2009, 2010]. The key quality of the method is its ability to simultaneously retrieve gas concentrations and particle scattering properties of the atmosphere using an efficient radiative transfer (RT) model [Hasekamp and Butz, 2008]. Particle scattering properties are effectively parameterized by a single spherical particle type characterized through its total column number density  $N_s$ , the size distribution parameter  $\alpha_s$ , the height distribution parameter  $z_s$ , and a fixed-value refractive index ( $1.400 - i \times 0.003$ ). Thereby, the particle number density size distribution follows a power-law  $\propto r^{-\alpha_s}$  with  $r$  the particle radius. The height distribution of particle optical thickness is a Gaussian function of center height  $z_s$ .

[8] Molecular absorption lines of O<sub>2</sub> and CO<sub>2</sub> are modeled by a spectroscopic model that includes line-mixing as well as collision-induced-absorption by O<sub>2</sub> [Tran and Hartmann, 2008; Lamouroux *et al.*, 2010]. Absorption of CH<sub>4</sub> and the interfering absorber H<sub>2</sub>O is modeled by feeding HITRAN 2008 [Rothman *et al.*, 2009] spectroscopic parameters into a Voigt lineshape model. Solar Fraunhofer lines are represented through an empirical linelist (G. Toon, JPL, personal communication, 2011). For nadir-view, surface reflection is assumed Lambertian, while for ocean-glint view, a wind-speed driven Cox-and-Munk surface model [Cox and Munk, 1954] is combined with a Lambertian albedo slope.

[9] Here, we run the RT model in scalar mode, i.e., we neglect polarization of radiation and only calculate the total radiance (Stokes parameter  $I$ ) backscattered to the satellite observer. Given the RT modeled and the measured radiances, an inverse method based on Phillips-Tikhonov regularization in combination with the L-curve method estimates the retrieval parameters. We exploit radiances in the 4 windows covering the O<sub>2</sub>A-band ([12920,13195]  $\text{cm}^{-1}$ ), a weakly absorbing CO<sub>2</sub> band ([6170,6278]  $\text{cm}^{-1}$ ), a CH<sub>4</sub> band ([6045,6138]  $\text{cm}^{-1}$ ), and a strongly absorbing CO<sub>2</sub> band ([4806,4896]  $\text{cm}^{-1}$ ). Retrieval parameters are the 12-layer vertical profiles of the CO<sub>2</sub> and CH<sub>4</sub> column number densities, the total column number density of the interfering absorber H<sub>2</sub>O, the scattering parameters  $N_s$ ,  $\alpha_s$ , and  $z_s$ , a 2nd order polynomial for surface albedo per window, and spectral shift parameters per window. Further, we retrieve a constant offset that adds to the radiance in the O<sub>2</sub>A-band window in order to effectively account for uncorrected non-linearity of the analogue-to-digital converter and contributions from plant fluorescence [Frankenberg *et al.*, 2011].

[10] The target quantities  $X_{\text{CO}_2}$  and  $X_{\text{CH}_4}$  are calculated by summing the respective column number densities over the 12 retrieval layers and dividing by the dry air column. The latter is calculated from the ECMWF meteorological support. Except for calibration purposes (section 4), we do not retrieve the dry air column or a related quantity such as surface pressure.

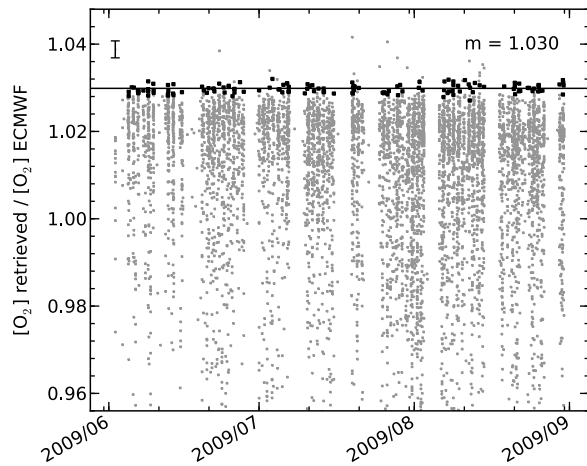
### 4. Calibrating O<sub>2</sub>

[11] Our retrieval method relies on accurate modeling of the O<sub>2</sub>A-band which provides a large part of the information content on atmospheric scattering properties. Therefore, we first investigate whether our model of the O<sub>2</sub>A-band is consistent with the measurements before aiming at  $X_{\text{CO}_2}$  and  $X_{\text{CH}_4}$  retrievals.

[12] To this end, we require TANSO-FTS O<sub>2</sub>A-band spectra that are not affected by aerosol and cloud scattering, and thus only depend on O<sub>2</sub> absorption and surface reflection. Ocean-glint observations allow for selecting such spectra. We set up a simplified version of our method, that retrieves the O<sub>2</sub> total column number density ( $[\text{O}_2]$ ) and the near surface windspeed from ocean-glint observations of the O<sub>2</sub>A-band ([12920,13195]  $\text{cm}^{-1}$ ) alone. The simplified forward model only accounts for Rayleigh scattering by molecules but neglects scattering by particles. For ocean glint observations, the latter assumption leads always to an underestimation of the retrieved  $[\text{O}_2]$  due to unaccounted lightpath shortening if scattering particles are present in the atmosphere. Overestimation of retrieved  $[\text{O}_2]$  does not occur since scattering induced lightpath enhancement typically requires a reflection at the ground which is inefficient due to low reflectivity of the ocean surface for off-glnt angles [Aben *et al.*, 2007].

[13] Figure 1 illustrates the ratio of retrieved  $[\text{O}_2]$  to the ECMWF  $[\text{O}_2]$  estimate for TANSO-FTS ocean-glnt measurements between June 1, 2009, and August 31, 2009. A cloud of low  $[\text{O}_2]$  retrievals is bound by a sharp 'upper edge'. Low-biased  $[\text{O}_2]$  relates to scattering effects as explained above. The 'upper edge' consists of scenes where the particle load is low and scattering effects are negligible. If the retrieval model of the O<sub>2</sub>A-band was consistent with the measurements,  $[\text{O}_2]$  retrievals along the 'upper edge' should be in agreement with the ECMWF estimate. However, we find a

<sup>1</sup>Auxiliary materials are available in the HTML. doi:10.1029/2011GL047888.



**Figure 1.** Ratio of  $[O_2]$  retrieved from TANSO-FTS ocean-glint observations to  $[O_2]$  calculated from ECMWF meteorological input. We show all individual retrievals (grey) between June 1, 2009, and August 31, 2009, after some basic quality filtering. Data contributing to the ‘upper edge’ (black dots) are used to fit a constant  $m$  (solid line). The ‘upper edge’ is defined such that  $\sim 32\%$  of the data above the fitted constant deviate by more than the 1-sigma noise error from the fit. For clarity, only a typical errorbar representing the 1-sigma noise error of the satellite retrievals is given in the upper left corner.

scaling factor of 1.030 between retrieved  $[O_2]$  and ECMWF  $[O_2]$  as determined by a least-squares fit of a constant to the ‘upper edge’.

[14] The origin of the detected scaling factor remains unclear so far but is most likely attributable to spectroscopic uncertainties. Irrespective of the actual origin of the inconsistency, we scale the  $O_2$  absorption cross sections in the  $O_2A$ -band by a factor 1.030 to make our  $O_2A$ -band model consistent with the observations.

## 5. Validating CO<sub>2</sub> and CH<sub>4</sub>

[15] For validation of our  $X_{CO_2}$  and  $X_{CH_4}$  retrievals from TANSO-FTS, we consider roughly the first year of satellite operation between April 2009 and July 2010. We focus on satellite observations in the vicinity of 6 TCCON sites in North-America (Lamont, 36.6°N 97.5°W; Park Falls, 45.9°N 90.3°W), Australia (Darwin, 12.4°S 130.9°E; Wollongong, 34.4°S 150.9°E), and Europe (Bialystok, 53.2°N 23.0°E; Orleans, 48.0°N 2.1°E). At each TCCON site, a ground-based sun-viewing FTS provides highly accurate estimates of  $X_{CO_2}$  and  $X_{CH_4}$  calibrated to the WMO standard [Deutscher et al., 2010; Messerschmidt et al., 2010; Wunch et al., 2010]. Unavailability of validation data is typically due to local cloud cover omitting direct sun-viewing and due to periods of instrument maintenance.

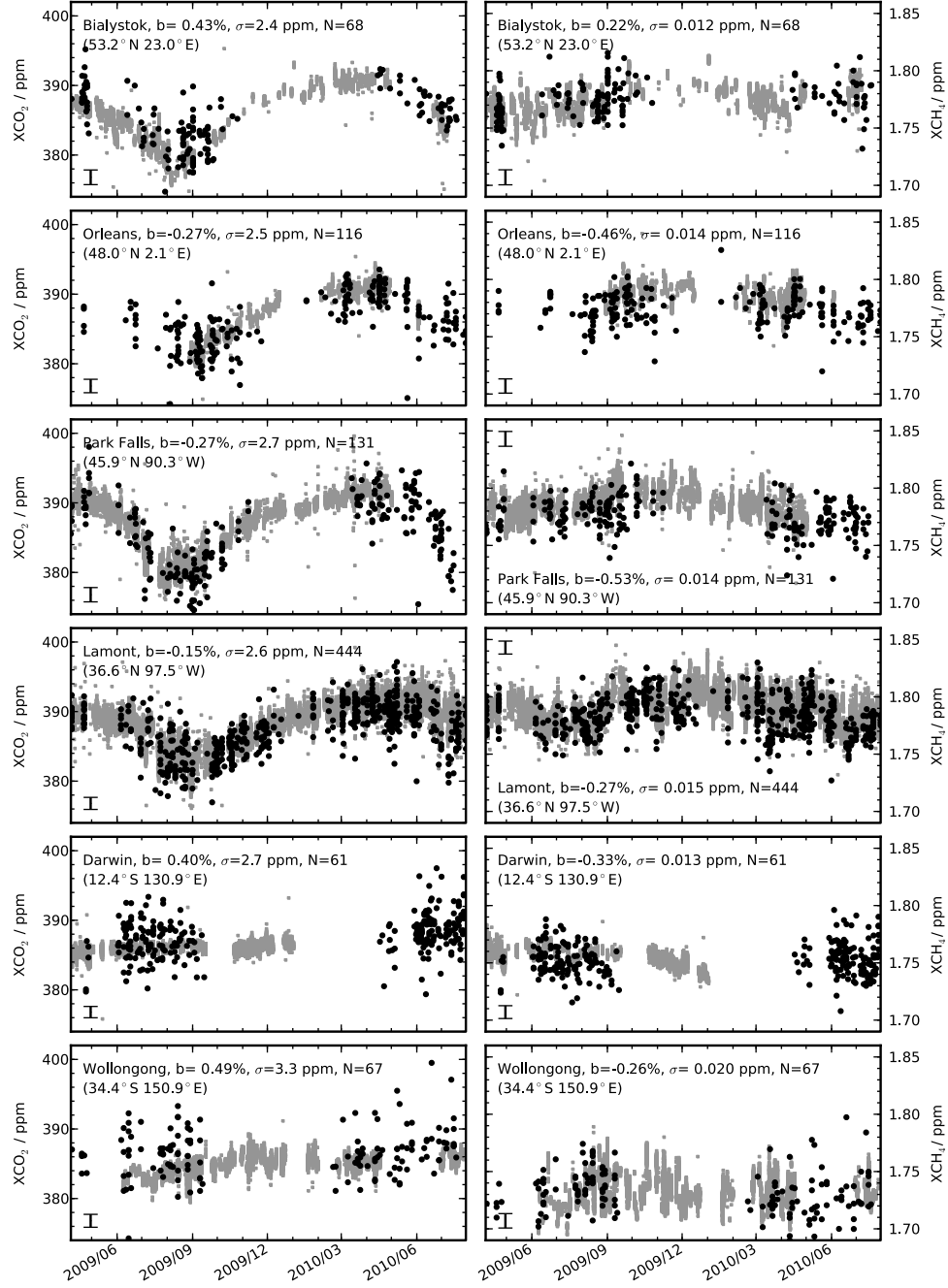
[16] We process TANSO-FTS spectra which are recorded in nadir-view over land within 5° latitude/longitude radius of the considered TCCON stations. Observations are *a priori* filtered according to several criteria such as cloudiness of the scene, instrumental error flags, extreme viewing geometry, surface roughness, and signal-to-noise. *A posteriori*, we reject bad quality fits and difficult scattering scenes based on

a ‘scattering’ criterion  $\tau_s \times 1/\alpha_s \times z_s [\text{km}] > 0.3$  where the filter threshold 0.3 is determined empirically and  $\tau_s$  is the retrieved particle optical thickness (at the  $O_2A$ -band) (see auxiliary material for details).

[17] Figure 2 compares the time series of  $X_{CO_2}$  and  $X_{CH_4}$  retrieved from TANSO-FTS to the validation data provided by TCCON. The diurnal range of TCCON data reflects the variability of air mass transport and source/sink processes as well as the effect of measurement errors [Keppel-Aleks et al., 2011]. For calculating the bias and scatter of the satellite retrievals per station, a temporal coincidence criterion is applied (maximum 2 h mismatch between satellite and ground-based observations). Among the 6 validation sites, the overall bias of the  $X_{CO_2}$  satellite retrievals is  $-0.05\%$  with a station-to-station variability of 0.37% (standard deviation of the bias). The  $X_{CH_4}$  retrievals exhibit an overall bias of  $-0.30\%$  and a station-to-station variability of 0.26%.  $X_{CH_4}$  retrievals at Bialystok seem exceptionally high in comparison to the other stations. Excluding the Bialystok record from the statistics reduces the station-to-station variability of the  $X_{CH_4}$  satellite record to 0.12% with an average bias of  $-0.34\%$ . As a measure of the scatter in our satellite retrievals, we calculate the root-mean-square deviation of the bias-corrected satellite record from the colocated TCCON data. The average scatter of  $X_{CO_2}$  and  $X_{CH_4}$  amounts to 2.8 ppm and 0.015 ppm among the 6 validation sites which is roughly twice the combined noise error estimated by the satellite and ground-based retrievals. The comparison for Wollongong reveals a large scatter of the TANSO-FTS retrievals for both target species which could be related to rough surface topography in Wollongong’s vicinity. Slight errors in the geolocation of the TANSO-FTS footprint could cause errors in assumed surface elevation and surface pressure and thus map into  $X_{CO_2}$  and  $X_{CH_4}$  errors. In general, statistics and conclusions are most robust for the Lamont site since data availability is high throughout the year. For the other stations, the applied filtering causes significant gaps in the satellite record which hinder the statistical analysis.

[18] In comparison to a retrieval method that entirely neglects particle scattering, our refined method in particular yields a reduced scatter of the  $X_{CO_2}$  and  $X_{CH_4}$  retrievals and provides the ‘scattering’ criterion that efficiently screens outliers (see auxiliary material). The calibration of the  $O_2A$ -band absorption cross sections as proposed in section 4 substantially affects the retrieved scattering parameters. Among the validation ensemble, the retrieved scattering optical thickness (at the  $O_2A$ -band) is found reduced from on average 0.17 without  $O_2$  scaling to 0.09 with  $O_2$  scaling. The latter retrievals agree reasonably well with colocated ground-based measurements made by the Atmospheric Radiation Measurement (ARM) program at Lamont (see auxiliary material). The average scattering layer height is found increased from 3.3 km without  $O_2$  scaling to 5.4 km with  $O_2$  scaling. Generally, omitting the  $O_2$  scaling results in a longer lightpath assumed by the retrieval method. Consequently, the retrieved  $X_{CO_2}$  and  $X_{CH_4}$  as shown in Figure 2 is lower by on average 0.21% and 0.27%, respectively, if  $O_2$  scaling is omitted. Station-to-station variability and scatter are only slightly affected.

[19] In general, the satellite retrievals capture the temporal and spatial patterns observed in the validation data well. In particular, the Northern hemisphere seasonal cycle of  $X_{CO_2}$



**Figure 2.** Time series of (left) XCO<sub>2</sub> and (right) XCH<sub>4</sub> retrievals from TANSO-FTS (solid black dots) in comparison to ground-based validation data (grey) provided by 6 TCCON stations (top to bottom ordered according to latitude). All validation data that pass the TCCON quality filters are shown. TANSO-FTS retrievals are *a priori* and *a posteriori* filtered as described in section 5. All valid TANSO-FTS retrievals within 5° latitude/longitude radius of the respective TCCON stations are depicted irrespective of the temporal mismatch between the ground-based and satellite measurements. For clarity, only a typical error bar representing the noise error of the satellite retrievals is given in the lower or upper left corner of each panel. Beside the name and the geolocation of the validation site, the panel legends quote the average bias  $b$  between TANSO-FTS retrievals and TCCON data, the root-mean-square deviation  $\sigma$  of the bias-corrected satellite record from the validation data, and the number  $N$  of data pairs considered for calculating these quantities.  $N$  is smaller than the plotted number of TANSO-FTS retrievals since a 2 h temporal coincidence criterion is applied in addition to the spatial criterion.

with the late summer minimum and the spring maximum is clearly discernible for Bialystok, Orleans, Park Falls, and Lamont. The seasonal cycle at Park Falls reveals the largest amplitude among the Northern hemisphere sites. As expected, the Southern hemisphere stations Darwin and Wollongong lack a pronounced X<sub>CO<sub>2</sub></sub> seasonal cycle. The X<sub>CH<sub>4</sub></sub> retrievals confirm the ~0.05 ppm difference between Northern and Southern hemisphere abundances. Seasonal patterns are less distinct for X<sub>CH<sub>4</sub></sub> than for X<sub>CO<sub>2</sub></sub> but variations such as a rapid increase of X<sub>CH<sub>4</sub></sub> in late summer 2009 at Lamont are observable in TANSO-FTS and TCCON data.

## 6. Conclusion

[20] We process GOSAT observations between April 2009 and July 2010 by a method that allows for the simultaneous retrieval of X<sub>CO<sub>2</sub></sub>, X<sub>CH<sub>4</sub></sub>, and parameters representing the amount, the size, and the height of scattering particles in the atmosphere. Comparison of the inferred X<sub>CO<sub>2</sub></sub> and X<sub>CH<sub>4</sub></sub> to validation measurements at 6 ground-based TCCON sites around the world indicates that our satellite retrievals exhibit a residual bias and a station-to-station variability of the bias on the order of a few tenths of a percent for both, X<sub>CO<sub>2</sub></sub> and X<sub>CH<sub>4</sub></sub>. The scatter of our GOSAT retrievals is well below 1%. This is a substantial improvement compared to earlier GOSAT validation efforts [Yoshida *et al.*, 2011; Morino *et al.*, 2011]. The improved quality of our X<sub>CO<sub>2</sub></sub> and X<sub>CH<sub>4</sub></sub> retrievals allows for unambiguously identifying source/sink signals such as the seasonal cycle and its amplitude for X<sub>CO<sub>2</sub></sub> and the interhemispheric gradient for X<sub>CH<sub>4</sub></sub> without any data averaging. Constraining surface fluxes by our X<sub>CO<sub>2</sub></sub> and X<sub>CH<sub>4</sub></sub> GOSAT records is the logical next step.

[21] **Acknowledgments.** Access to GOSAT data was granted through the 2nd GOSAT research announcement jointly issued by JAXA, NIES, and MOE. Funding of this research came from the Dutch User Support Program under project GO-2005/064 and GO-AO/21 (DS), from ESA's CCI on GHGs and the European Commission's 7th framework program under grant agreement 218793 (SG), from DFG's Emmy-Noether program under project RemoteC BU2599/1-1 (AB). ECMWF ERA Interim analyses are provided through [http://data-portal.ecmwf.int/data/d/interim\\_daily/](http://data-portal.ecmwf.int/data/d/interim_daily/). GTOPO30 is available from the U.S. Geological Survey through the Earth Resources Observation and Science (EROS) Center ([http://eros.usgs.gov/#/Find\\_Data/Products\\_and\\_Data\\_Available/gtopo30\\_info](http://eros.usgs.gov/#/Find_Data/Products_and_Data_Available/gtopo30_info)). CarbonTracker data are provided by NOAA ESRL, Boulder, Colorado, USA from the Web site at <http://carbontracker.noaa.gov>. TM4 fields have been made available through Jan-Fokke Meirink, Royal Netherlands Meteorological Institute (KNMI). US funding for TCCON comes from NASA's Terrestrial Ecology Program (NNX08A186G), the Orbiting Carbon Observatory Program (NAS7-03001), the DOE/ARM Program and the Atmospheric CO<sub>2</sub> Observations from Space Program. Some of the research described in this paper was performed at the Jet Propulsion Laboratory, California Institute of Technology, under a contract with the NASA. We acknowledge the support of the European Commission within the 6th Framework Program through the Integrated Infrastructure Initiative IMECC and the Integrated Project GEOMon. We thank AeroMeteo Service, (Bialystok, Poland) and the RAMCES team at LSCE (Gif-sur-Yvette, France) for maintaining the Bialystok and Orleans FTS sites and providing station logistics.

[22] The Editor thanks two anonymous reviewers for their assistance in evaluating this paper.

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